

09/936,692

3

In the claims:

1. (Currently Amended) An ionomer composite composition having improved physical properties, the composite consisting essentially of:

a glass material containing at least one of divalent cations and multivalent cations;

and

at least one copolymer, comprising:

at least one hydrophilic monomer containing acid functional groups adapted to react with the at least one of divalent cations and multivalent cations to form ionic crosslinks among polymer chains, the hydrophilic monomer present in an amount sufficient to impart a desired degree of aqueous solubility to the copolymer; and

at least one hydrophobic monomer present in an amount sufficient to impart a desired degree of structural stability to the composite composition when exposed to an aqueous environment;

wherein the at least one hydrophilic monomer and the at least one hydrophobic monomer are present in a predetermined ratio of hydrophilic monomer to hydrophobic monomer, wherein the higher the amount of hydrophilic monomer in the ratio the more water soluble the composition, and the higher the amount of hydrophobic monomer in the ratio the more water insoluble the composition.

2. (Currently Amended) The ionomer composite composition as defined in claim 1 wherein the glass material further contains fluoride, and wherein the composite composition provides ~~substantially continuous~~ fluoride release.

3. (Original) The ionomer composite composition as defined in claim 1 wherein the glass material is selected from the group consisting of SiO₂, Al₂O₃, AlF₃, CaF₂, NaF, Na₃AlF₆, AlPO₄, and mixtures thereof.

4. (Original) The ionomer composite composition as defined in claim 1 wherein a weight ratio of glass material to copolymer ranges between about 10:90 and about 90:10.

09/936,692

4

5. (Original) The ionomer composite composition as defined in claim 4 wherein the weight ratio of glass material to copolymer ranges between about 40:60 and about 85:15.

6. (Original) The ionomer composite composition as defined in claim 1 wherein the at least one hydrophilic monomer is a monomer containing unsaturated carboxylic acid.

7. (Currently Amended) The ionomer composite composition as defined in claim 6 wherein the unsaturated carboxylic acid is selected from the group consisting of: acrylic acid, methacrylic acid, 4-vinylbenzoic acid, crotonic acid, oleic acid, elaidic acid, itaconic acid, maleic acid, fumaric acid, acetylenedicarboxylic acid, tricarbollylic acid, sorbic acid, linoleic acid, linolenic acid, eicosapentenoic acid, anhydrides of the acids, hydrophilic derivatives of the unsaturated carboxylic acids, and mixtures thereof.

8. (Currently Amended) The ionomer composite composition as defined in claim 7 wherein the hydrophilic monomer is selected from the group consisting of organic acids having at least one of sulfonic acid and phosphonic acid replacement of the carboxyl group of the unsaturated carboxylic acids, hydrophilic derivatives of the organic acids, and mixtures thereof.

9. (Original) The ionomer composite composition as defined in claim 1 wherein the at least one hydrophobic monomer is unsaturated.

10. (Currently Amended) The ionomer composite composition as defined in claim 9 wherein the at least one hydrophobic monomer is selected from the group consisting of acrylates, methacrylates, ethylenes, propylenes, tetra-fluoroethylene, styrenes, vinyl chloride, vinylidene chloride, vinyl acetate, acrylonitrile, 2,2-bis[4-(2-hydroxy-3-methacryloyloxy-propyloxy)-phenyl] propane (Bis-GMA), ethyleneglycol dimethacrylate (EGDMA), tri-ethyleneglycol dimethacrylate (TEGDMA), bis(2-methacryly-oxyethyl) ester of isophthalic acid (MEI), bis(2-meth-acrylyoxyethyl) ester of terephthalic acid (MET), bis(2-methacrylyoxyethyl) ester of phthalic acid (MEP), 2,2-bis(4-methacrylyoxy phenyl) propane (BisMA), 2,2-bis[4-(2-methacrylyloxyethoxy) phenyl] propane (BisEMA), 2,2-bis[4-(3-methacrylyloxy-propoxy) phenyl] propane (BisPMA), hexafluoro-1,5-pentanediol dimethacrylate (HFPDMA), bis-(2-

09/936,692

5

methacrylyloxyethoxy-hexafluoro-2-propyl) benzene [Bis(MEHFP) ϕ], 1,6-bis(methacrylyloxy-2-ethoxycarbonylamino)-2,4,4-tri-methylhexan (UEDMA), spiro orthocarbonates, hydrophobic derivatives of the hydrophobic monomers, and mixtures thereof.

11. (Original) The ionomer composite composition as defined in claim 1 wherein a ratio of the hydrophilic monomer to the hydrophobic monomer is between about 1:99 and about 99:1.

12. (Currently Amended) The ionomer composite composition as defined in claim 1 wherein the at least one copolymer is selected from the group consisting of poly(methyl methacrylate-methacrylic acid), poly(methyl acrylate-acrylic acid), poly(methyl methacrylate-acrylic acid), poly(ethyl acrylate-acrylic acid), poly(ethyl methacrylate-methacrylic acid), poly(butyl acrylate-acrylic acid), poly(ethylene-acrylic acid), poly(ethylene-methacrylic acid), poly(acrylonitrile-maleic anhydride), poly(butadiene-acrylonitrile-acrylic acid), poly(butadiene-maleic acid), poly(butadiene-maleic anhydride), ~~poly(acrylamide-acrylic acid)~~, ~~poly(2-hydroxyethyl methacrylate-methacrylic acid)~~, poly(propylene-acrylic acid), poly(propylene-ethylene-acrylic acid), poly(vinyl chloride-vinyl acetate-maleic acid), and mixtures thereof.

13. (Original) The ionomer composite composition as defined in claim 1 wherein the composite further includes as a minor constituent a polymer selected from the group consisting of PMMA, polycarbonates, polyethylenes, polyamides, PEEK polymers, epoxies, and mixtures thereof.

14. (Original) The ionomer composite composition as defined in claim 2 wherein the composite further includes a minor constituent selected from the group consisting of pigments, surfactants, adhesion enhancers, fluoride releasing enhancers, bioactive agents, and mixtures thereof.

15. (Original) The ionomer composite composition as defined in claim 2 wherein the glass material consists of 25.10 wt% SiO₂, 37.45 wt% Al₂O₃, and 37.45 wt% CaF₂.

09/936,692

6

16. (Original) The ionomer composite composition as defined in claim 1 wherein the at least one copolymer is poly(methyl methacrylate-co-methacrylic acid) (PMMA-MAA).

17. (Original) The ionomer composite composition as defined in claim 16 wherein the ratio of PMMA to MAA is 80:20.

18. (Currently Amended) An ionomer composite composition having improved physical properties and useful for dental applications, the composite consisting essentially of:

a glass material containing at least one of divalent cations and multivalent cations;

and

at least one copolymer, comprising:

at least one hydrophilic monomer containing acid functional groups adapted to react with the at least one of divalent cations and multivalent cations to form ionic crosslinks among polymer chains, the hydrophilic monomer present in an amount sufficient to impart a desired degree of aqueous solubility to the copolymer; and

at least one hydrophobic monomer present in an amount sufficient to impart a desired degree of structural stability to the composite composition when exposed to an aqueous environment;

wherein the at least one hydrophilic monomer and the at least one hydrophobic monomer are present in a predetermined ratio of hydrophilic monomer to hydrophobic monomer, wherein the higher the amount of hydrophilic monomer in the ratio, the more water soluble the composite composition, and the higher the amount of hydrophobic monomer in the ratio, the more water insoluble the composite composition; and

wherein the composite composition is adaptable for use in dental applications ranging from those requiring water soluble compositions, to intermediary materials to partially soluble compositions, to those requiring water insoluble compositions.

19. (Currently Amended) The ionomer composite composition as defined in claim 18 wherein the glass material further contains fluoride, and wherein the composite composition provides ~~substantially continuous~~ fluoride release.

09/936,692

7

20. (Original) The ionomer composite composition as defined in claim 19 wherein the glass material is selected from the group consisting of SiO_2 , Al_2O_3 , AlF_3 , CaF_2 , NaF , Na_3AlF_6 , AlPO_4 , and mixtures thereof.

21. (Original) The ionomer composite composition as defined in claim 20 wherein the weight ratio of glass material to copolymer ranges between about 40:60 and about 85:15.

22. (Currently Amended) The ionomer composite composition as defined in claim 21 wherein the at least one hydrophilic monomer is an unsaturated carboxylic acid selected from the group consisting of: acrylic acid, methacrylic acid, 4-vinylbenzoic acid, crotonic acid, oleic acid, elaidic acid, itaconic acid, maleic acid, fumaric acid, acetylenedicarboxylic acid, tricarbollylic acid, sorbic acid, linoleic acid, linolenic acid, eicosapentenoic acid, anhydrides of the acids, hydrophilic derivatives of the unsaturated carboxylic acids, and mixtures thereof.

23. (Currently Amended) The ionomer composite composition as defined in claim 22 wherein the at least one hydrophilic monomer is selected from the group consisting of organic acids having at least one of sulfonic acid and phosphonic acid replacement of the carboxyl group of the unsaturated carboxylic acids, hydrophilic derivatives of the organic acids, and mixtures thereof.

24. (Currently Amended) The ionomer composite composition as defined in claim 22 wherein the at least one hydrophobic monomer is unsaturated and is selected from the group consisting of acrylates, methacrylates, ethylenes, propylenes, tetra-fluoroethylene, styrenes, vinyl chloride, vinylidene chloride, vinyl acetate, acrylonitrile, 2,2-bis[4-(2-hydroxy-3-methacryloyloxy-propyloxy)-phenyl] propane (Bis-GMA), ethyleneglycol dimethacrylate (EGDMA), tri-ethyleneglycol dimethacrylate (TEGDMA), bis(2-methacryly-oxyethyl) ester of isophthalic acid (MEI), bis(2-meth-acrylyoxyethyl) ester of terephthalic acid (MET), bis(2-methacrylyoxyethyl) ester of phthalic acid (MEP), 2,2-bis(4-methacryloxy phenyl) propane (BisMA), 2,2-bis[4-(2-methacryloxyethoxy) phenyl] propane (BisEMA), 2,2-bis[4-(3-methacryloxy-propoxy) phenyl] propane (BisPMA), hexafluoro-1,5-pentanediol dimethacrylate (HFPDMA), bis-(2-methacrylyloxyethoxy-hexafluoro-2-propyl) benzene

09/936,692

8

[Bis(MEHFP) ϕ], 1,6-bis(methacryloyloxy-2-ethoxycarbonylamino)-2,4,4-trimethylhexan (UEDMA), spiro orthocarbonates, hydrophobic derivatives of the hydrophobic monomers, and mixtures thereof.

25. (Original) The ionomer composite composition as defined in claim 24 wherein a ratio of the hydrophilic monomer to the hydrophobic monomer is between about 1:99 and about 99:1.

26. (Currently Amended) The ionomer composite composition as defined in claim 18 wherein the at least one copolymer is selected from the group consisting of poly(methyl methacrylate-methacrylic acid), poly(methyl acrylate-acrylic acid), poly(methyl methacrylate-acrylic acid), poly(ethyl acrylate-acrylic acid), poly(ethyl methacrylate-methacrylic acid), poly(butyl acrylate-acrylic acid), poly(ethylene-acrylic acid), poly(ethylene-methacrylic acid), poly(acrylonitrile-maleic anhydride), poly(butadiene-acrylonitrile-acrylic acid), poly(butadiene-maleic acid), poly(butadiene-maleic anhydride), ~~poly(acrylamide-acrylic acid), poly(2-hydroxyethyl methacrylate-methacrylic acid),~~ poly(propylene-acrylic acid), poly(propylene-ethylene-acrylic acid), poly(vinyl chloride-vinyl acetate-maleic acid), and mixtures thereof.

27. (Original) The ionomer composite composition as defined in claim 18 wherein the composite further includes as a minor constituent a polymer selected from the group consisting of PMMA, polycarbonates, polyethylenes, polyamides, PEEK polymers, epoxies, and mixtures thereof.

28. (Original) The ionomer composite composition as defined in claim 19 wherein the composite further includes a minor constituent selected from the group consisting of pigments, surfactants, adhesion enhancers, fluoride releasing enhancers, bioactive agents, and mixtures thereof.

29. (Original) The ionomer composite composition as defined in claim 19 wherein the glass material consists of 25.10 wt% SiO₂, 37.45 wt% Al₂O₃, and 37.45 wt% CaF₂.

09/936,692

9

30. (Original) The ionomer composite composition as defined in claim 19 wherein the at least one copolymer is poly(methyl methacrylate-co-methacrylic acid) (PMMA-MAA).

31. (Original) The ionomer composite composition as defined in claim 30 wherein the ratio of PMMA to MAA is 80:20.

Please add the following new claims:

32. (New) A method of making an ionomer composite composition, comprising the steps of:

mixing a glass material containing at least one of divalent cations and multivalent cations and at least one copolymer, the copolymer comprising:

at least one hydrophilic monomer containing acid functional groups adapted to react with the at least one of divalent cations and multivalent cations to form ionic crosslinks among polymer chains, the hydrophilic monomer present in an amount sufficient to impart a desired degree of aqueous solubility to the copolymer; and

at least one hydrophobic monomer present in an amount sufficient to impart a desired degree of structural stability to the composite composition when exposed to an aqueous environment; and

varying a ratio of the at least one hydrophilic monomer to the at least one hydrophobic monomer, wherein the higher the amount of hydrophilic monomer in the ratio, the more water soluble the composition, and the higher the amount of hydrophobic monomer in the ratio, the more water insoluble the composition.

33. (New) The method of claim 32 wherein an application for which the ionomer composite composition is used ranges from one application requiring water soluble compositions, to partially soluble compositions, to one requiring water insoluble compositions.

34. (New) The method of claim 32 wherein the glass material further contains fluoride, and wherein the composite composition provides fluoride release.

09/936,692

10

35. (New) The method of claim 32 wherein the glass material is selected from the group consisting of SiO_2 , Al_2O_3 , AlF_3 , CaF_2 , NaF , Na_3AlF_6 , AlPO_4 , and mixtures thereof.

36. (New) The method of claim 32 wherein a weight ratio of glass material to copolymer ranges between about 10:90 and about 90:10.

37. (New) The method of claim 36 wherein the weight ratio of glass material to copolymer ranges between about 40:60 and about 85:15.

38. (New) The method of claim 32 wherein the at least one hydrophilic monomer is a monomer containing unsaturated carboxylic acid.

39. (New) The method of claim 38 wherein the unsaturated carboxylic acid is selected from the group consisting of: acrylic acid, methacrylic acid, 4-vinylbenzoic acid, crotonic acid, oleic acid, elaidic acid, itaconic acid, maleic acid, fumaric acid, acetylenedicarboxylic acid, tricarbollylic acid, sorbic acid, linoleic acid, linolenic acid, eicosapentenoic acid, anhydrides of the acids, hydrophilic derivatives of the unsaturated carboxylic acids, and mixtures thereof.

40. (New) The method of claim 32 wherein the hydrophilic monomer is selected from the group consisting of organic acids having at least one of sulfonic acid and phosphonic acid replacement of the carboxyl group of the unsaturated carboxylic acids, hydrophilic derivatives of the organic acids, and mixtures thereof.

41. (New) The method of claim 32 wherein the at least one hydrophobic monomer is unsaturated.

42. (New) The method of claim 41 wherein the at least one hydrophobic monomer is selected from the group consisting of acrylates, methacrylates, ethylenes, propylenes, tetra-fluoroethylene, styrenes, vinyl chloride, vinylidene chloride, vinyl acetate, acrylonitrile, 2,2-bis[4-(2-hydroxy-3-methacryloyloxy-propyloxy)-phenyl] propane (Bis-GMA), ethyleneglycol dimethacrylate (EGDMA), tri-ethyleneglycol dimethacrylate (TEGDMA), bis(2-methacryly-

09/936,692

11

oxyethyl) ester of isophthalic acid (MEI), bis(2-meth-acrylyoxyethyl) ester of terephthalic acid (MET), bis(2-methacrylyoxyethyl) ester of phthalic acid (MEP), 2,2-bis(4-methacryloxy phenyl) propane (BisMA), 2,2-bis[4-(2-methacrylyloxyethoxy) phenyl] propane (BisEMA), 2,2-bis[4-(3-methacrylyloxy-propoxy) phenyl] propane (BisPMA), hexafluoro-1,5-pentanediol dimethacrylate (HFPDMA), bis-(2-methacrylyloxyethoxy-hexafluoro-2-propyl) benzene [Bis(MEHFP) ϕ], 1,6-bis(methacrylyloxy-2-ethoxycarbonylamino)-2,4,4-tri-methylhexan (UEDMA), spiro orthocarbonates, hydrophobic derivatives of the hydrophobic monomers, and mixtures thereof.

43. (New) The method of claim 32 wherein a ratio of the hydrophilic monomer to the hydrophobic monomer is between about 1:99 and about 99:1.

44. (New) The method of claim 32 wherein the at least one copolymer is selected from the group consisting of poly(methyl methacrylate-methacrylic acid), poly(methyl acrylate-acrylic acid), poly(methyl methacrylate-acrylic acid), poly(ethyl acrylate-acrylic acid), poly(ethyl methacrylate-methacrylic acid), poly(butyl acrylate-acrylic acid), poly(ethylene-acrylic acid), poly(ethylene-methacrylic acid), poly(acrylonitrile-maleic anhydride), poly(butadiene-acrylonitrile-acrylic acid), poly(butadiene-maleic acid), poly(butadiene-maleic anhydride), poly(propylene-acrylic acid), poly(propylene-ethylene-acrylic acid), poly(vinyl chloride-vinyl acetate-maleic acid), and mixtures thereof.

45. (New) The method of claim 32 further comprising the step of adding, as a minor constituent, a polymer selected from the group consisting of PMMA, polycarbonates, polyethylenes, polyamides, PEEK polymers, epoxies, and mixtures thereof.

46. (New) The method of claim 32 further comprising the step of adding a minor constituent selected from the group consisting of pigments, surfactants, adhesion enhancers, fluoride releasing enhancers, bioactive agents, and mixtures thereof.

47. (New) The method of claim 34 wherein the glass material consists of 25.10 wt% SiO₂, 37.45 wt% Al₂O₃, and 37.45 wt% CaF₂.

09/936,692

12

48. (New) The method of claim 32 wherein the at least one copolymer is poly(methyl methacrylate-co-methacrylic acid) (PMMA-MAA).

49. (New) The method of claim 48 wherein the ratio of PMMA to MAA is 80:20.